## 410 Rec'd PCT/PTO 2 4 JUN 1999

#### PATENT IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

| In re Application of:   | )                              |
|---|--------------------------------|
| HANS-PETER ESSER, ET AL.  | ) Examiner: Not Assigned       |
| International Appl. No. PCT/EP97/04662  | )<br>)                         |
| U.S. Serial No.: 09/254,625   | ) Group Art Unit: Not Assigned |
| Filed: March 11, 1999   | ) Atty. Dkt. No.: 16202.160    |
| For: PROCESS FOR THE PRODUCTION OF<br>BIOLOGICALLY DEGRADABLE<br>ALIPHATIC POLYESTER AMIDE SOLU-<br>TIONS | )<br>)<br>)<br>)               |
| BOX PCT<br>Commissioner of Patents and Trademarks<br>Washington, DC 20231                                 |                                |
| Sir:  |                                |

#### **LETTER**

Submitted herewith are copies of all papers filed on May 12, 1999 in compliance with the Notification of Missing Requirements Under 35 U.S.C. 371 in the United States

Designated/Elected Office (DO/EO/US) dated April 12, 1999.

We are enclosing a copy of check No. 13263 in the amount of \$195.00 and a copy of our PTO post card.

It is believed that the Notification of Missing Requirements dated 5/24/99 was mailed in error.

It is also believed that no fee is due for this submission. Should that determination be incorrect, please debit Account No. 50-0548 and notify the undersigned.

In view of the above, the applicant respectfully submits that all documents necessary to for perfecting the filing date of March 11, 1999 have been filed, and that a filing receipt be issued.

Respectfully submitted

Joseph W. Berenato Registration No. 30,546 Attorney for Applicant

Liniak, Berenato, Longacre & White, LLC 6550 Rock Spring Drive Suite 240
Bethesda, Maryland 20817 (301) 896-0600

6550 ROCK SPRING DR., SUITE 240 BETHESDA, MD 20817-1132 PH. (301) 896-0600



13263

5/12/99

PAY TO THE ORDER OF.

Commissioner of Patents

\*\*195.00

One Hundred Ninety-Five and 00/100\*

**DOLLARS** 

Commissioner of Patents Washington, DC 20231

**MEMO** 

16202.160 Surcharge and Translation fee

"O13263" #255071981

**AUTHORIZED SIGNATURE** 

LINIAK, BERENATO, LONGACRE & WHITE / A LIMITED LIABILITY COMPANY

Commissioner of Patents

05/12/99

Bill #16202.160

🖴 security features included. Details on Back. 🗟

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Chevy Chase Bank L & B 16202.160 Surcharge and Translation fee

195.00

THE PATENT OFFICE DATE STAMP HEREON ACKNOWLEDGES RECEIPT OF THE BELOW INDICATED DOCUMENT:

SERIAL NUMBER:-09/254,625 APPLICANTS:- Hans-Peter Esser

Subm. of Missing Pts.

- 2) Deci.
- Prel. Amdit.

- 4) Trans. English, Trans. Declaration 5) Check No. 13263 for \$ 195.00 6) Assign., Assign Trans. and Check for \$40 JWBldmg 16202.160 5-12-99 Courier



Patent and Trade Tk Office

Address: ASSISTAN SIONER FOR PATENTS

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|--|---|-----------------------------------|--|---|
| 19/25/1/25   | No of the last of |                                   | n. D.C. 20231                          | · /   |
| U.S. APPLICATION NO.   |   | FIRST NAMED                       | APPLICANT                              | ATTY, DOCKET NO.  |
| 09/254,625.  | ESSER   |                                   | H /                                    | 16202.160   |
| •  | •   |                                   | DATELL                                 | IATIONAL APPLICATION NO.  |
| 1  | 561°  | CEIVED                            | PCT                                    | /EP97/04662   |
| JOSEPH W BERENATO I  | 11  | FPS(A PT).                        |  | DATE PRIORITY DATE  |
| LINIAK BERENATO LON  | GACRE & WHITE   |                                   | I.A. FILING                            | DATE PRIORITY DATE  |
| 6550 ROCK SPRING DR  | IVE ;   | 7 1200 L                          | 08/27/                                 | 97 09/12/96   |
| SUITE 240<br>BETHESDA MD 20817   | :   | $\mathcal{L}$                     |  |   |
| 1  | L.  | riak, Berenato,<br>ngaore & White | DATE MAILED:                           | 04/12/99  |
| NOTIFICATION OF MIS  | SING DEOI HREM  | ENTS UNDER                        | 35 U.S.C.                              | 371 IN THE UNITED   |
|  |   |                                   |  |   |
| 1. The following items have been   | submitted by the applica  | nt or the IB to the               | United States                          | Patent and Trademark  |
| Office no 17 a Designated U  | JMCE (37 CFK 1.777),  |                                   |  | •   |
| Fin Elected Off  | ice (37 CFR 1.495):   |                                   |  |   |
| U.S. Basic National Fee. Copy of the international a   | nolication in:  |                                   |  |   |
| a non-English  | language.   |                                   | •                                      |   |
| The Prolish.   | •   |                                   |  |   |
| Translation of the internation   | onal application into Englishments for DO/FO/IIS.   | usn.                              |  |   |
| Oath or Declaration of inve  | nenis.  |                                   | •                                      |   |
| To manufaction of Article 10 of  | mendments into English.   |                                   | · * 6                                  |   |
| - Destining  | er Bramination Kenon i  | n English and its                 | Annexes, il any                        | lish  |
| Translation of Annexes to  | the International Prelimin  | and                               | Report mix Line                        | va ,  |
| Preliminary amendment(s) Information Disclosure Sta  | med   | and                               |  | <u> </u>  |
| Assignment document.   | •   |                                   |  |   |
| Power of Attorney and/or   | Change of Address.  | •                                 |  |   |
| Substitute specification file  | d   | <del></del> •                     | -                                      |   |
| Statement Claiming Small   |   |                                   |  |   |
| Eopy of the International  | Search Report 🖂 and co  | pies of the referen               | ices cited therei                      | n.  |
| Other: 2. The following items MUST be  |   |                                   |  |   |
|  |   |                                   |  | 6°  |
| Translation of the appli   | cation into English. Not  | e a processing fee                | will be require                        | d if submitted  |
| —  | to 70 or 30 months from   | the priority date.                |  | · ·   |
| The current trans  | station is defective for the  | e reasons indicate                | d on me auacuc                         | A Modes of Datasers   |
| Translation.  b. Processing fee for pro-   | viding the translation of t   | he application and                | l/or the Annexe                        | s later that the  |
|  |   |                                   |  |   |
| - who or declaration of  | the inventors, in complia   | ince with 31 Cer                  | . 1.431(a) and (                       | b), identifying the application   |
|  |   |                                   |  | nd (b) for the reasons indicat  |
| / =  | DCT/DC/EC/017   |                                   |  |   |
| rad. Surcharge for providir  | ng the oath or declaration  | later that the app                | propriate 20 or                        | 30 months from the  |
| priority date (37 CFR  |   |                                   |  | g any required multiple   |
| <ol> <li>Additional claim fees of \$</li></ol>   | Annlicant must subm   | it the additional c               | laim fees or car                       | acel the additional claims for  |
| which fees are due (37 CFR 1.4   | 92(g)). See attached PT   | 0-875.                            |  |   |
|  |   | A A DOVE MI                       | ST BE SUBMI                            | TTED WITHIN ONE   |
| ALL OF THE ITEMS SET F<br>MONTH FROM THE DATE  | OF THIS NOTICE OR   | BY   21 OR                        | 31 MONTHS                              | FROM THE PRIORITY   |
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| The time period set above may  | be extended by filing a   | petition and fee fo               | r extension of ti                      | me under the provisions of 3  |
| CFR 1.136(a).  |   |                                   | ragista responsa se se area.<br>Tanàna |   |
| 4. Translation of the Annexes  | MUST be submitted no  | later that the time               | period set abov                        | e or the annexes will be  |
| 4. Translation of the Annexes  | e will be required if subm  | litted later than 30              | months from the                        | ne priority date.   |
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| 1.494(d)) or 30 (37 CFR 1.49   | 2(9)) moints trom me by   | orny, care                        | 11 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 |   |
| Applicant is reminded that any   | communication to the U  | nited States Patent               | and Trademari                          | t Office must be mailed to in   |
| address given in the heading a   | nd melude me o.s. appa  | ·                                 |  | الله<br>التي التي الشيرية المراجعة المستراجية المراجعة المراجعة المراجعة المراجعة المراجعة المراجعة المراجعة المراجعة |
| _ £ 4L.  | s notice MIIS   | T he retur                        | rned with                              | this response.  |
| A copy of the  | D INVINCE IN COLUMN   | fective Translation               | 216/11                                 | · //  |
| Enclosed: PCT/DO/EO/9  | . –   | 181                               | M/パーパー<br>hone: (703) - 3              | 1818181   |
| FORM PCT/DO/EO/905 (De   | cember 1997)  | Telep                             | ער (ניטי) :Suone                       | BALLER  |
|  |   |                                   |  |   |

# U.S. APPLICATION NO.



## UNITED STATES DF TIMENT OF COMMERCE Patent and Trademark Tice Address: ASSISTANT COMMISSIONER FOR PATENTS Box PCT Washington, D.C. 20231

|   | -765 0             |                       |                        |
|---|--------------------|-----------------------|------------------------|
| U.S. APPLICATION NO. 097.254,625                        |                    | FIRST NAMED APPLICANT | ATTY. DOCKET NO.       |
| 007204,620  | ESSER              | Н                     | 16202.160              |
| IOCEDU II DED   | 5611               | INTERNAT              | TIONAL APPLICATION NO. |
| JOSEPH W BERENATO III<br>LINIAK BERENATO LONGACRE & WHI | O LONGACRE & WHITE | P                     | CT/EP97/04662          |
| 6550 ROCK SPRIN   | •                  | I.A. FILING DA        | TE PRIORITY DATE       |
| BETHESDA MD 20817                                       | 317                | 087:                  | 27/97 09/12/97         |
|   |                    | DATE MAILED           | 05/24/99               |

| JUSEPH W BERENATO III  | DOT (FROM ALL)   |
|--|--|
| LINIAK BERENATO LONGACRE & WHITE   | PCT/EP97/04662   |
| 6000 RUCK SPRING DRIVE   | LA STUNG DUTC  |
| SUITE 240  | I.A. FILING DATE PRIORITY DATE   |
| BETHESDA MD 20817  | 08/27/97 09/12/96  |
|  | 1 22, 12, 30   |
|  | 05/24/99 DATE MAILED:  |
| NOTIFICATION OF MISSING PROLUDENCE AND ADDRESS OF THE PROPERTY |  |
| NOTIFICATION OF MISSING REQUIREMENTS UNDER   | R 35 U.S.C. 371 IN THE UNITED  |
| STATES DESIGNATED/ELECTED OFF  | TCF MO/FO/He)  |
| 1. The following items have been submitted by the applicant or the IB to the Office as [7] a Designated Office (37 CFR 1 404)  | e United States Patent and Trademark   |
| Ta Designated Office (37 CFR 1.494).   | RECEIVED   |
|  | REAL COLUMN  |
| Copy of the international application in:  |  |
| a non-English language.  | MAY 28 1999  |
| English.   |  |
| Translation of the international application into English.   | Liniak, Berenato,  |
| Oath or Declaration of inventors(s) for DO/EO/US.  | Longacre & White   |
| Copy of Article 19 amendments.   | Longadio di  |
| Translation of Article 19 amendments into English.   |  |
| The International Preliminary Examination Report in English and its  | Annexes if any   |
| I ranslation of Annexes to the International Preliminary Examination I   | Report into English  |
| Preliminary amendment(s) filed and   | report into English.   |
| Information Disclosure Statement(s) filed and  | <del></del> •  |
| Assignment document.   | <del></del> •  |
| Power of Attorney and/or Change of Address.  |  |
| Substitute specification filed   |  |
| Statement Claiming Small Entity Status.  | •  |
| Priority Document.   |  |
| Copy of the International Search Report (X) and copies of the reference Other:   | es cited therein.  |
|  |  |
| 2. The following items MUST be furnished within the period set forth below acceptance under 35 U.S.C. 371:   | in order to complete the requirements for  |
| a. Translation of the application into English. Note a processing fee w  | will be required to a to the   |
| later than the appropriate 20 or 30 months from the priority date.   | our de required it submitted   |
| Translation is defective for the reasons indicated of  | on the attached Notice of Defend   |
| i ansiation.   |  |
| b. Processing fee for providing the translation of the application and/or  | the Annexes later that the   |
| appropriate 20 or 30 months from the priority date (37 CFR 1 402/f   | A)   |
| (X) c. Oath or declaration of the inventors, in compliance with 37 CFR 1 4   | 197(a) and (b) identifying the analysis  |
| of the international application number and international filing date  | •  |
| The current oath or declaration does not comply with 37 CFR  | 1.497(a) and (b) for the reasons indicated   |
| on the attached religion (CO) (CO) (CO) (CO)   |  |
| d. Surcharge for providing the oath or declaration later that the appropriate priority date (37 CFR 1.492(e)).   | riate 20 or 30 months from the   |
|  |  |
| 3. Additional claim fees of \$ as a large entity small entity dependent claim fee, are required. Applicant must submit the additional claim which fees are due (37 CFR 1.492(a)). See a march of PRO 2005.   | ry, including any required multiple  |
| which fees are due (37 CFR 1.492(g)). See attached PTO-875.  | lees of cancel the additional claims for   |
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| ALL OF THE ITEMS SET FORTH IN 2(a)-2(d) AND 3 ABOVE MUST B   | E SUBMITTED WITHIN ONE   |
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| RESULT IN ABANDONMENT.   |  |
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| · L 1 · · · · · · · · · · · · · · · · ·  | ided by the appropriate 20 (25 GP)   |
| .494(d)) or 30 (37 CFR 1.495(d)) months from the priority date.  | ded by the appropriate 20 (37 CFR  |
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| pplicant is reminded that any communication to the United States Patent and T  | rademark Office must be mailed to the  |
| ddress given in the heading and include the U.S. application no. shown above.  | (37 CFR 1.5)   |
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| A copy of this notice MUST be returned   | with this res  |
| Notice of Defective Translation //   | 11:00  |
| P10-8/5  | aun Will   |
| 7/EO/905 (December 1997) Telephone: (7   | (03) 305   |
| ·  | - Control of the Cont |

## PATENT IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

| In re Application of                   | )                          |
|--|----------------------------|
| Hans-Peter Esser                       | ) Art Unit: Not Assigned   |
| Serial No. 09/254,625                  | ) Examiner: Not Assigned   |
| International Appl. No. PCT/EP97/04662 | )                          |
| Filing Date: March 11, 1999            | )                          |
|  | ) Atty. Dkt. No. 16202.160 |
| For: PROCESS FOR THE PRODUCTION        | )                          |
| OF BIOLOGICALLY DEGRADABLE             | )                          |
| ALIPHATIC POLYESTER AMIDE              | )                          |
| SOLUTIONS                              | )                          |
| Box PCT                                |                            |
| Commissioner of Patents                | •                          |
| and Trademarks                         |                            |

#### **SUBMISSION OF MISSING PARTS**

Dear Sir:

Washington, D.C. 20231

In response to the Notification of Missing Requirements Under 35 U.S.C. 371 in the United States Designated/Elected Office (DO/EO/US) mailed April 12, 1999, (copy attached), enclosed are the following:

- 1) Declaration for Patent Application referencing the application entitled
  PROCESS FOR THE PRODUCTION OF BIOLOGICALLY DEGRADABLE
  ALIPHATIC POLYESTER AMIDE SOLUTIONS, referencing Attorney Docket No.
  16202.160;
- 2) Check No.13263 in the amount of \$195.00 to cover the cost of the surcharge and English translation fee Small Entity Status has been established;
  - 3) English translation and Translator's Declaration; and
  - 4) Preliminary Amendment.

The undersigned attorney of record asserts that the application filed in the Patent and Trademark Office is the application which the inventors executed by signing the Oath or Declaration.

Should any other charges be due, then please debit Account No. 50-0548, and notify the undersigned.

In view of the above, the applicant respectfully submits that all documents necessary for perfecting the filing date of March 11, 1999 have been filed, and that a filing receipt be issued.

Date: May 12, 1999

Joseph W. Berenato, III Registration No. 30,546 Agent for Applicant

Respectfully submitted,

Liniak, Berenato, Longacre & White, LLC 6550 Rock Spring Drive, Suite 240 Bethesda, Maryland 20817 (301) 896-0600

## PATENT IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

| In re Application of                   | )                          |
|--|----------------------------|
| Hans-Peter Esser                       | ) Art Unit: Not Assigned   |
| Serial No. 09/254,625                  | ) Examiner: Not Assigned   |
| International Appl. No. PCT/EP97/04662 | )<br>)                     |
| Filing Date: March 11, 1999            | )                          |
| E DROOFIGE FOR THE DROPHICTION         | ) Atty. Dkt. No. 16202.160 |
| For: PROCESS FOR THE PRODUCTION        | <b>)</b>                   |
| OF BIOLOGICALLY DEGRADABLE             |                            |
| ALIPHATIC POLYESTER AMIDE              | •                          |
| SOLUTIONS                              | )                          |
| Box PCT                                |                            |
| Commissioner of Patents                |                            |
| and Trademarks                         | *                          |
| Washington D.C. 20231                  |                            |

#### PRELIMINARY AMENDMENT

#### Dear Sir:

Prior to the initial examination of the above-identified patent application, please amend the application as follows:

#### IN THE CLAIMS:

Claim 3, line 1, delete "or 2".

Claim 4, line 1, delete "one of Claims 1 through 3" and insert -- Claim 1--.

Claim 5, line 1, delete "one of Claims 1 through 4" and insert -- Claim 1--.

Claim 6, line 1, delete "one of Claims 1 through 5" and insert -- Claim 1--.

Claim 7, line 1, delete "one of Claims 1 through 6" and insert -- Claim 1--.

Claim 9, line 1, delete "one of Claims 7 or 8" and insert -- Claim 7--.

Claim 10, line 1, delete "one of Claims 7 through 9" and insert -- Claim 7--.

Claim 11, line 1, delete "one of Claims 7 through 10" and insert -- Claim 7--.

Claim 12, line 1, delete "one of Claims 7 through 11" and insert --Claim 7--.

Claim 13, line 1, delete "one of Claims 1 through 12" and insert --Claim 1--.

Claim 16, line 1, delete "one of Claims 1 through 12" and insert --Claim 1--.

Claim 17, line 1, delete "one of Claims 1 through 12" and insert --Claim 1--.

#### **REMARKS**

The preceding Amendment has been made in order to amend multiple dependencies in the claims. Applicant asserts that all claims are in condition for examination. It is believed that no fee is due for this submission. Should that determination be incorrect, please debit Account No. 50-0548 and notify the undersigned.

Date: May 12, 1999

Registration No. 30,546 Agent for Applicant

Respectfully submitted,

Joseph W. Berenato, III

Liniak, Berenato, Longacre & White, LLC 6550 Rock Spring Drive, Suite 240 Bethesda, Maryland 20817 (301) 896-0600



#### Nicholas Hartmann 828 N. Broadway, Suite 506 Milwaukee, WI 53202

(414) 271-4890 fax: (414) 271-4892 polyglot@execpc.com

Technical and scientific translation - German and French into English

### Declaration

I, Nicholas Hartmann, translator, having an office at 828 N. Broadway, Suite 506, Milwaukee, WI, 53202, declare that I am well acquainted with the English and German languages and that the appended document is a true and faithful translation of:

International Patent Application numbered WO 98/11153, PCT/EP97/04662, and entitled:

"Verfahren zum Herstellen von Lösungen biologisch abbaubarer Kunststoffe, insbesondere aliphatischer Polyesteramide"

All statements made herein are to my own knowledge true, and all statements made on information and belief are believed to be true; and further, these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the document.

Date 4/2(89

Nicholas Hartmann

#### PROCESS FOR THE PRODUCTION OF BIOLOGICALLY DEGRADABLE ALIPHATIC POLYESTER AMIDE SOLUTIONS

The present invention concerns a method for the manufacture of solutions of biodegradable plastics, in particular of aliphatic polyester amides, and the use of the resulting solution for the manufacture of films and the coating of substrates made of metal, paper, wood, plastic, ceramic, and foodstuffs.

Plastics are widely used in households, commerce, and industry, for example as shaped elements, films, and coatings. Disposal thereof after use, however, represents an increasing problem. In recent years, biodegradable plastics have therefore also been developed.

Polyester amides based on natural amino acids are known from Polym. Bull. 28 (1992) 301-307. They are manufactured by way of a complex protective-group technique, since natural amino acids in combination with hydroxycarboxylic acids are generally involved; this is very cumbersome. In addition, these polymers have absolutely no mechanical properties which are necessary for the manufacture of useful objects.

Further biodegradable polyester amides made of lactic acid, diamines, and dicarboxylic acid dichlorides are disclosed in US Patents 4,343,931 and 4,529,792.

Japanese Patents 79 113 593 and 79 109 594 disclose biodegradable polymers made from caprolactone and caprolactam. The polyester amides just mentioned are, however, complex to manufacture.

A further polyester amide is disclosed in European Patent Application EP 641 817. The polyester amide described therein can be processed thermoplastically, and is biodegradable. It has a melting point of at least 75<sub>i</sub>C, and the weight proportion of the ester structures is between 30 and 70%, and the proportion of amide structures between 70 and 30%. The aforesaid polymer has good mechanical properties, but processability is very difficult. Shaped elements can be manufactured from the polymer only in the mass. Solutions, for example in ethanol, are not stable, and quickly result in decomposition of the polymer.

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It is the object of the present invention to manufacture solutions of biodegradable polyester amides so as to impart simplified and improved processability to them.

The subject matter of the present invention is a method for manufacturing solutions of biodegradable plastics, in particular of aliphatic polyester amides, which is characterized in that the aliphatic polyester amide is added to a solvent mixture containing

A) a C1-C4 alcohol;

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- B) a C1-C6 ketone; and/or
- C) an aromatic carboxylic acid or a salt thereof.

It has been found, surprisingly, that biodegradable aliphatic polyester amides can be readily dissolved in the solvent mixture according to the present invention which contains components A, B and/or C. After only a few minutes, the polymer swells in the solution and dissolves. The dissolution rate can optionally be increased by mechanical actions such as agitation.

The resulting solution is stable for several days with no observable decomposition of the polymer structure.

According to a preferred embodiment, the method comprises the following steps:

- a) the plastic is placed in a vessel;
- b) the solvent mixture is added to the vessel until the plastic is covered by the solvent mixture;
- c) the vessel is sealed and the plastic and solvent mixture are allowed to stand until the plastic has softened and swollen;
- d) the softened and swollen plastic is mechanically comminuted and the resulting emulsion is preferably filtered.

During the swelling operation, it may be advantageous to add further solvent in order to accelerate the swelling operation or effect further swelling of the plastic, if the

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solvent mixture originally added to the vessel has been completely absorbed by the plastic.

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In order to increase the absorption surface area of the plastic in this context, it may be opportune to comminute the plastic mechanically during the swelling operation so as thereby to accelerate the process.

In order to obtain a clear solution, the softened and swollen plastic is preferably filtered; the filtered-out solids can be added to a new batch of plastic + solvent mixture.

According to an alternative embodiment, powdered plastic is introduced into the solvent with continuous agitation, so that it dissolves immediately and a coating can be made.

Methanol and/or ethanol are preferably used as the C1-C4 alcohols of component A; it is preferred for environmental reasons to use methanol and ethanol obtained from plant-based raw materials. The solvent mixture contains the C1-C4 alcohol preferably in a quantity of 70 to 98.9 wt%, in particular in a quantity of 90 to 98.9 wt%.

Acetone and butanone (methyl ethyl ketone) have proven particularly suitable as the C1-C6 ketone. The ketone is present in the solvent preferably in a quantity from 0.1 to 5 wt%, preferably from 0.1 to 2 wt%.

Benzoic acid and its derivatives, i.e. compounds in which the aromatic ring is the substituent, have proven particularly successful as aromatic carboxylic acids. Benzoates are preferably used, denatonium benzoate being particularly preferred. Component C is usually present in the solvent mixture in a quantity of 0.01 to 5 ppm.

One polyester amide that is preferably used is made up of aliphatic monomers in which the weight proportion of the ester structure is between 30 and 70% and the proportion of the amide structure is between 70 and 30%. To allow the polymer to be used outdoors, i.e. even in sunlight, the polyester amide should have a melting point of at least 75 iC.

Polyester amides that have proven suitable are, in particular, those described in

European Patent Application EP 0 641 817.

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The average molecular weight (HW as determined by gel chromatography in n-cresol against a polystyrene standard) is from 10,000 to 300,000, preferably 20,000 to 150,000.

The polyester amides preferably used can be obtained in a manner known per se, for example by mixing the amide-forming and ester-forming starting components and then polymerizing them. Synthesis can also be accomplished by the "polyamide" method by stoichiometrically mixing the starting components, optionally with the addition of water and subsequent removal of water from the reaction mixture; and by the "polyester" method, by adding an excess of diol with branching of the acid groups, following by rebranching or reamidization of those esters. In the second variant method, excess glycol is also distilled off in the water.

The ester and amide segments are arranged purely statistically, governed fundamentally by the synthesis conditions. It is also possible, however, to use polyester amides in which the monomers are distributed as longer segments in the polymer molecule.

The following are used, for example, as monomers for the manufacture of the polyester amides which are preferably used:

Dialcohols such as ethylene glycol, 1,4-butanediol, 1,3-butanediol, 1,6-hexanediol, diethylene glycol, etc.; and/or dicarboxylic acids such as oxalic acid, succinic acid, adipic acid and their lower alkyl esters; and/or hydroxycarboxylic acids and lactones, such as caprolactone, etc.; and/or amino alcohols such as ethanolamine, propanolamine, etc.; and/or cyclic lactams such as a-caprolactam and laurolactam; and/or v-amino carboxylic acids such as aminocaproic acid, etc.; and/or mixtures (1:1 salts) of dicarboxylic acids such as adipic acid, succinic acid, etc. and diamines such as hexamethylenediamine, diaminobutane, etc.

Hydroxyl- or acid-terminated polyesters having molecular weights between 200

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and 10,000 can also be used as the ester-forming component.

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The resulting polyester amides can also contain 0.1 to 5 wt%, preferably 0.1 to 2 wt%, of so-called branching agents. Compounds of this kind can be, for example, trifunctional alcohols such as trimethylolpropane or glycerol, tetrafunctional alcohols such as pentaerythrite, trifunctional carboxylic acids such as citric acid. Incorporation of such components increases the melt viscosity of the polyester amides. The biodegradability of these materials is not, however, impeded.

The solvent mixture used according to the present invention can additionally contain, as well as the aforementioned components A, B, and C, further constituents which improve the solubility of the polymers and may stabilize the solution. The mixture can also contain water in a quantity up to 30 wt%, preferably between 0.1 and 10 wt%.

The films manufactured according to the present invention offer a much wider application spectrum for biodegradable polyester amides than the pure substances.

For example, it is possible to cast films from the solutions. Clear, elastic films are obtained, which can be manufactured in any desired thickness and can be used, for example, as compostable trash bags or milk films.

The films can contain any desired fillers; care should be taken that the compostability of the polymers is not impaired by such additives. Examples of fillers are talc, CaSO4 (for example, gypsum which is produced by flue gas desulfuration), compost, peat, garden mold, etc. The last-named fillers in particular make it possible to use the biodegradable polymers in agriculture and horticulture.

A further possible application of the solution obtained according to the present invention is utilization for coating substrates made of metal, paper, wood, plastic, ceramic, and foodstuffs. One possibility is use as a protective coating for metal substrates and glass as a protective coating during transport. In addition, for example, paper or board can be coated, so that the mechanical properties of board and paper and their resistance to moisture and water are enhanced, but the paper can be environmentally recycled after use. Coating can be accomplished, for example, with a dip method, by brush application, or

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with a spray method. The films formed in each case can be pulled off in their entirety very quickly and without leaving a residue.

A further application of the solutions obtained according to the present invention is as an adhesive. For this, the solutions in concentrated form are applied onto the surfaces or points to be joined, and the surfaces are pressed together after a short exposure time which allows the solvent mixture to evaporate.

#### Example 1:

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10 g of the polymer was placed in a beaker. 300 ml of a solvent mixture comprising 94 wt% ethyl alcohol, 2 wt% methyl ethyl ketone, 0.1 wt% denatonium benzoate, and water to make 100%, was added thereto and allowed to stand for three days. Swelling of the polymer was observed after a few hours. After two days the polymer was completely dissolved, yielding a clear, low-viscosity solution.

The polymer used was BAK 1095 (commercial product of Bayer AG, Leverkusen). BAK 2195 can also be used.

#### 15 Example 2:

300 g of the polymer was placed in a beaker. The solvent mixture was added and was allowed to stand for 24 hours without heat, in a sealed vessel in which a vacuum had been drawn, until an increase in volume and a color change in the plastic had been noted.

The plastic was again covered with solvent, and then allowed to stand for approximately 24 hours in the sealed beaker.

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This operation was repeated until an approximately threefold increase in volume had been noted, and the plastic was almost clear. The surface of the plastic was then sufficiently soft that mechanical comminution with a simple stirrer was possible. This operation was also repeated several times, further solvent being added as necessary for

dilution, until a low-viscosity solution was obtained. This was pressed through a fine-mesh filter to remove the solids still present in the emulsion.

The plastic used was BAK 1095. BAK 2195 can also be used.

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#### Claims

| 1   | 1.  | A method for the manufacture of solutions of biodegradable plastics, in   |  |
|-----|---|---|--|
| 2   | particular of aliphatic polyester amides, wherein the aliphatic polyester amide is added to |   |  |
| 3   | a solvent mix   | xture containing  |  |
| 4 . | A)  | a C1-C4 alcohol;  |  |
| 5 . | B)  | a C1-C6 ketone; and/or  |  |
| 6   | C)  | an aromatic carboxylic acid or a salt thereof.                            |  |
| 1   | 2.  | The method as defined in Claim 1, wherein methanol and/or ethanol are     |  |
| 2   | used as the C   | C1-C4 alcohol.  |  |
| 1   | 3.  | The method as defined in one of Claims 1 or 2, wherein acetone and/or     |  |
| 2 - | methyl ethyl  | ketone are used as the ketone.  |  |
| 1   | 4.  | The method as defined in one of Claims 1 through 3, wherein the aromatic  |  |
| 2   | carboxylic ac   | cid is benzoic acid.  |  |
| 1 . | 5.  | The method as defined in one of Claims 1 through 4, wherein the polyester |  |
| 2   | amide is a co   | polymer based on aliphatic monomers and has a melting point of at least   |  |
| 3   | 75;C, and the   | e weight proportion of the ester structure is between 30 and 70%, and the |  |
| 4   | proportion of   | f the amide structure is between 70 and 30%.                              |  |
| 1   | 6.  | The method as defined in one of Claims 1 through 5, wherein the solvent   |  |
| 2   | mixture cont  | ains water in a quantity up to 30 wt%.                                    |  |
| 1   | 7.  | The method as defined in one of Claims 1 through 6, characterized by the  |  |

#### 2 following steps: a) the plastic is placed in a vessel; the solvent mixture is added to the vessel until the plastic is covered by the **b**) 5 solvent mixture; the vessel is sealed and the plastic and solvent mixture are allowed to 6 c) 7 stand until the plastic has swollen and softened; 8 the softened and swollen plastic is mechanically comminuted and the 9 resulting emulsion is preferably filtered. 1 8. The method as defined in Claim 7, wherein the swelling operation takes 2 place under vacuum. 9. 1 The method as defined in Claim 7 or 8, wherein solvent is added again at 2 least once while the plastic is swelling. 1 10. The method as defined in one of Claims 7 through 9, wherein the swelling 2 time is 2 to 60 hours. 1 11. The method as defined in one of Claims 7 through 10, wherein further 2 solvent is added during comminution of the swollen plastic. 1 12. The method as defined in one of Claims 7 through 11, wherein the solids 2 filtered out during filtration are added to a new batch of plastic + solvent mixture.

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Use of the solvent obtained as defined by one of Claims 1 through 12 for

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the manufacture of films.

1 14. The use as defined in Claim 13, wherein the films contain fillers.

1 15. The use as defined in Claim 14, wherein compost, peat, garden mold,

and/or CaSO4 are used as fillers.

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- 16. Use of the solution obtained as defined in one of Claims 1 through 12 for coating substrates made of metal, glass, paper, wood, plastic, ceramic, and foodstuffs.
- 17. Use of the solution obtained as defined in one of Claims 1 through 12 as an adhesive.

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#### [Modified sheets, IPEA/EP]

#### Claims

- 1. A method for the manufacture of solutions of biodegradable aliphatic polyester amides, wherein the aliphatic polyester amide is added to a solvent mixture containing
  - A) a C1-C4 alcohol;
  - B) a C1-C6 ketone; and/or
  - C) an aromatic carboxylic acid or a salt thereof.
- 2. The method as defined in Claim 1, wherein methanol and/or ethanol are used as the C1-C4 alcohol.
- 3. The method as defined in one of Claims 1 or 2, wherein acetone and/or methyl ethyl ketone are used as the ketone.
- 4. The method as defined in one of Claims 1 through 3, wherein the aromatic carboxylic acid is benzoic acid.
- 5. The method as defined in one of Claims 1 through 4, wherein the polyester amide is a copolymer based on aliphatic monomers and has a melting point of at least 75; C, and the weight proportion of the ester structure is between 30 and 70%, and the proportion of the amide structure is between 70 and 30%.
- 6. The method as defined in one of Claims 1 through 5, wherein the solvent mixture contains water in a quantity up to 30 wt%.

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- 7. The method as defined in one of Claims 1 through 6, characterized by the following steps:
  - a) the polyester amide is placed in a vessel;
- b) the solvent mixture is added to the vessel until the polyester amide is covered by the solvent mixture;
- c) the vessel is sealed and the polyester amide and solvent mixture are allowed to stand until the polyester amide has swollen and softened;
- d) the softened and swollen polyester amide is mechanically comminuted and the resulting emulsion is preferably filtered.
- 8. The method as defined in Claim 7, wherein the swelling operation takes place under vacuum.
- 9. The method as defined in Claim 7 or 8, wherein solvent is added again at least once while the polyester amide is swelling.
- 10. The method as defined in one of Claims 7 through 9, wherein the swelling time is 2 to 60 hours.
- 11. The method as defined in one of Claims 7 through 10, wherein further solvent is added during comminution of the swollen polyester amide.
- 12. The method as defined in one of Claims 7 through 11, wherein the solids filtered out during filtration are added to a new batch of polyester amide + solvent mixture.
  - 13. Use of the solvent obtained as defined by one of Claims 1 through 12 for

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the manufacture of films.

- 14. The use as defined in Claim 13, wherein the films contain fillers.
- 15. The use as defined in Claim 14, wherein compost, peat, garden mold, and/or CaSO4 are used as fillers.
- 16. Use of the solution obtained as defined in one of Claims 1 through 12 for coating substrates made of metal, glass, paper, wood, plastic, ceramic, and foodstuffs.
- 17. Use of the solution obtained as defined in one of Claims 1 through 12 as an adhesive.